AN INITIAL ASSESSMENT OF INFILTRATION MATERIAL SELECTION FOR SELECTIVE LASER SINTERED PREFORMS

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Abstract

High-temperature infiltration is an important process that is used to add strength to skeletal microstructures. In this study, particulate metal matrix composites (MMCs) are fabricated. MMCs are applied in a wide variety of industries, including military, aircraft, tooling and automotive. In this paper, various materials for infiltrating selective laser sintered (SLS) silicon carbide and titanium carbide preforms are considered based on fundamental knowledge of SLS and infiltration mechanics. Proposed infiltrant materials systems include an aluminum-silicon alloy infiltrant and a silicon carbide preform, ductile iron infiltrated into a titanium carbide preform, and commercially pure silicon infiltrated into a silicon carbide preform. The first two infiltrants are considered because they add ductility to the brittle silicon carbide or titanium carbide part, thus broadening the range of applications. They also will model a broader field of possible infiltrants, including magnesium and iron-based materials, (e.g., steel). Silicon is investigated because it adds strength to silicon carbide, is robust at high temperatures, and has a comparable coefficient of thermal expansion. Presented is a feasibility assessment of these systems based on infiltration theory.

Introduction

Particulate metal matrix composites are a significant technical material. Reinforcement material synthesis, matrix and reinforcement joining, and choice of matrix and reinforcement material are central parameters in this field [1]. In this research, selective laser sintering (SLS) will be used to fabricate a ceramic preform. Pressureless infiltration will be used to join the preform and matrix. Three materials systems will be used, including; titanium carbide (TiC) reinforcement with an iron matrix, silicon carbide (SiC) reinforcement with a silicon matrix, and a silicon carbide reinforcement with an aluminum matrix. Quality control issues include porosity control, agglomeration of the reinforcement, and poor interfacial bonding [1]. Gas pychnometry, scanning electron microscopy, and three-point bend testing will be used to assess these quality concerns.

The major significance of this work is using selective laser sintering as the process for fabricating the ceramic preform. SLS is a process that uses a rastering laser, directed by a computer-defined shape, to sinter powder particles into a compact structure [2]. In this study, preforms remain porous for infiltration; an indirect SLS method will be used [3]. A phenolic binder will be used to increase green strength of the preform. The binder also acts as a catalyst for the pressureless infiltration process. The binder will be burned out of the preform, leaving a carbon residue that draws silicon (either as an alloying element or as the matrix material) into the structure.

Due to SLS's functionality, a variety of materials can be processed, as can a broad range of geometrical shapes. In the past, preforms were pressed, die cast, or extruded. Using those forming processes, only very simple shapes were possible. SLS widens the array of shapes available, using infiltration as a post-processing step to increase functionality, thus broadening the assortment of applications while reducing manufacturing cost.

Background

Selective Laser Sintering

Selective Laser Sintering is the process used to create the preform, or reinforcement material. The first step in the process is to deposit a thin layer of powder across a piston that can be lowered in small increments. A computer program then directs a laser beam across the surface of the powder to be sintered. After that area of powder has been sintered, the piston lowers, and another layer of powder is deposited onto to the piston. Sintering multiple layers can form a three-dimensional shape in this manner.

Because the laser is on top of a small area of powder for only a very short period of time, that area is only heated a small amount. A few important processing parameters include spot size of the laser, scan spacing, scan speed, and powder bed and platform build temperature and heating cycle.

Pressureless Infiltration

Pressureless infiltration is a process where spontaneous infiltration of a liquid drawn through the pores of a solid by capillary forces occurs without the need for external pressure, meaning process conditions are met so that self-induced infiltration is thermodynamically favorable [1, 2, 4]. The liquid takes the place of the vapor from the pores and leaves behind a matrix of molten metal, as is the case in MMCs. This method is advantageous because it reduces process cost and minimizes shrinkage and distortion [4]. Infiltration parameters include infiltration time, temperature, wetting, viscosity of the liquid, atmosphere, alloy composition, pore size of the preform, size of reinforcement, and percent porosity in the preform [2, 4].

Time and Temperature:

The amount of time needed to infiltrate depends on the pore diameter, the liquid surface tension, and the liquid viscosity [5]. For material selection, the melting point of the infiltrant should be lower than that of the reinforcement. Processing temperature needs to be high enough to liquefy the infiltrant, but low enough to keep the preform solid.

Wetting:

Wetting of the liquid onto the solid is determined by the interfacial energies between the solid, liquid, and vapor, given by Young's equation:

$$\gamma_{sv} - \gamma_{sl} = \gamma_{lv} \cos \Theta$$

where γ_{sv} is the surface energy between the solid and the vapor, γ_{sl} is the surface energy between the solid and the liquid, γ_{lv} is the surface energy between the liquid and vapor, and Θ is the contact angle. Wetting occurs when $\Theta < 90°$, thus becoming favorable for pressureless infiltration. Wetting can be enhanced by a thermally activated chemical reaction at the solid-liquid interface [6]. Products made as a result of these chemical reactions may sometimes be unfavorable.

Alloying materials:
Alloying materials may give favorable products. Some alloying elements may react with surface layers on the reinforcement particulate, thus leaving uncontaminated particulate, allowing molten matrix material and the particulate to react.

Viscosity:
The viscosity of the infiltrant should be low enough to allow flow through the capillaries. An increase in solids loading increases the apparent viscosity. When the solids loading is above approximately 62%, flow ceases [7]. Alloying elements will vary viscosity, depending on their molecular or atomic weights; therefore, choosing an infiltrant and considering its alloying elements are essential.

Atmosphere, Pore diameter, and Reinforcement size:
An inert atmosphere may allow residual binder to be removed from the system in the flow of a noble gas. It can also take away other reactants and impurities from the alloy that may inhibit positive thermally activated chemical reactions. The pressure needed to draw a liquid into a preform is given by the Washburn Equation:

$$P = -4\,\gamma_{lv} \cos\theta/d$$

where d is the average pore diameter. The threshold pressure P* for spontaneous infiltration into a preform is

$$P^* = 6\lambda\,(-\gamma_{lv} \cos\theta)\,\Delta / (1-\Delta)\,D$$

where D is the particle size, λ is the ratio of the particle actual surface area to the surface area of a sphere of the same volume, and Δ is the relative density defined as the ratio of the part density to the material theoretical density. Setting P to be a smaller negative number than P*,

$$d \geq 2\,(1-\Delta)\,D / 3\,\lambda\,\Delta$$

If infiltration works against gravity, there is an equilibrium height balance between the wetting and gravitational forces,

$$H_{max} = 4\,\gamma_{lv} \cos\theta / dg\rho$$

Where g is the gravitational acceleration and ρ is the density [8].

Percent porosity:
A large percent porosity in the preform gives more volume for the matrix material to infiltrate. Retained porosity after infiltration may occur if the compaction during SLS creates voids within sintered particles. Here, the molten infiltrant may not reach the void because there is no path for wicking. Time, wetting, and viscosity of the infiltrant are variables that are pertinent to making

sure a dense infiltrated material is obtained. A superheated processing temperature increases fluidity, thus decreasing retained porosity [9].

Materials and Processing

TiC/Fe:

The three material systems chosen for this study are TiC reinforcement with an Fe matrix, SiC reinforcement with a Si matrix, and SiC reinforcement with an Al matrix. The first system, Ti/Fe is of interest because TiC is one of the toughest materials to be found, and in conjunction with Fe, will have better ductility characteristics. This material system may compete with the already established WC-Co markets [10]. Aside from WC-Co applications, other potential uses of this material include light armor for military use, wear parts, and cutting tools.

TiC and Fe have a high affinity for each other, therefore wetting and interfacial energies should be favorable during infiltration. Viscosity of the infiltrant will be dependent upon processing time and temperature (soak temperature of approximately 1100ºC). Argon and nitrogen will be used to create an inert atmosphere for processing [11]. Pore diameter and reinforcement size, and percent pre-infiltration porosity will be determined via SLS processing.

SiC/Si:

SiC's high resistance to thermal stress, excellent corrosion resistance in high temperature atmospheres, good wear resistance, and affordability make it useful in a number of applications, including wafer carriers, furnace components, electronic devices, grinding wheels, and gas turbine engine components. The reason Si is a good matrix material is because its coefficient of thermal expansion is close to that of SiC, thus creating a material good for thermal stresses.

Si and SiC will have favorable wetting, especially since there will be residual carbon left on the surface of the preform from binder burnout to attract the Si. Processing temperature will be held at approximately 1600ºC for a period of time to be determined. Argon and nitrogen will be used to create an inert atmosphere for processing [11]. Pore diameter, preform size, and percent pre-infiltration porosity will be the same as in the other two experiments.

In an oxidizing atmosphere, SiO_2 can form on the surface of the SiC, which can inhibit infiltration of molten Si into the preform. Capillary forces themselves are not enough for the infiltration to occur spontaneously. A chemical reaction is needed to reduce the solid-liquid infiltration energy to achieve successful pressureless infiltration. Over 1350°C, the reaction

$$2SiO_{2(s)} == 2SiO_{(g)} + O_{2(g)}$$

will occur in an oxidizing atmosphere and will react with nitrogen gas to form Si_3N_4 by the reaction

$$3SiO_{(g)} + 2N_{2(g)} == Si_3N_{4(s)} + 1.5O_{2(g)}$$

Silicon nitride formation during the heating process provides better wetting of molten Si on the SiC preform [3]. On the Si infiltrant material, formation of Si_3N_4 may also occur according to the reaction

$$3Si_{(s)} + 2N_{2(g)} == Si_3N_{4(s)}$$

Liquid Si infiltration will not occur if the Si_3N_4 wraps the Si infiltrant pellet. Two ways to keep these unwanted reactions from occurring are to minimize or eliminate oxygen from the process atmosphere and also to enclose the infiltrant reinforcement materials in a crucible [3].

SiC/Al:

SiC/Al metal matrix composites are one of the most commercially viable materials among the class of particulate metal matrix composites currently being researched for high thermal conductivity, and low coefficient of thermal expansion materials [12]. However, there are some concerns in processing these materials into an MMC. Two major problems that arise are poor wetting of SiC by molten aluminum due to a layer of aluminum oxide that covers the surface of the liquid aluminum, and the potential for unwanted reactions at the Al-SiC interface [Petch]. The first problem is inadequate wetting of SiC by aluminum. A layer of aluminum oxide may cover the molten aluminum, thus creating insufficient surface energies for a favorable contact angle. The way this is corrected is by using an alloy with Mg additions. The Mg acts as a surfactant and reduces the aluminum oxide on the surface of the aluminum, as well as the reinforcement-matrix interface. Magnesium has a low vapor pressure, thus the presence of nitrogen in the atmosphere helps to keep the magnesium back into the molten aluminum [13]. The nitrogen also positively affects the liquid-vapor surface tension as to increase wetting.

The second problem is the potential for unwanted intermetallics that may occur at the Al-SiC interface. At the interface, aluminum carbide may form by the reaction

$$4Al_{(s)} + 3SiC_{(s)} == Al_4C_{3(s)} + 3Si(s)$$

This reaction results in dissolution of the reinforcing preform, thus weakening the composite. Another possible product to form during SiC-Al contact is

$$4Al_{(s)} + 4SiC_{(s)} == Al_4SiC_{4(s)} + 3Si(s)$$

Both products have detrimental effects in that they compromise the SiC preform by using it to form other products. An increase in the Si in the aluminum alloy decreases the incidence of aluminum carbide formation [13]. However, too much Si in the system can cause precipitation of Si crystals. These crystals are relatively brittle, and compromise the mechanical properties of the material [Pickard]. An approximate eutectic alloy of Al-Si will be used. Processing temperature will be approximately 600ºC for a time to be determined. Pore diameter, part size, and percent pre-infiltrated porosity will be the same as the prior two experiments.

Conclusion

Experiments using the three infiltrant material/reinforcement systems as variables will be performed. Infiltration techniques vary for the material system used. An inert atmosphere will be used during the TiC/Fe infiltration process. In the SiC/Si system, an inert atmosphere will be applied and enclosing the reinforcement material will be considered. Magnesium will be utilized as an alloying element in the aluminum matrix for the SiC/Al system. The amount of silicon in the alloy will also be considered. Characterization of the MMC materials will include gas pychnometry, scanning electron microscopy, and three-point bend testing.

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